

Health Consultation

EXPOSURE INVESTIGATION

HERCULANEUM LEAD SMELTER SITE

HERCULANEUM, JEFFERSON COUNTY, MISSOURI

EPA FACILITY ID: MOD006266373

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Division of Health Assessment and Consultation

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In addition, consultations may recommend additional public health actions, such as conducting health surveillance activities to evaluate exposure or trends in adverse health outcomes; conducting biological indicators of exposure studies to assess exposure; and providing health education for health care providers and community members. This concludes the health consultation process for this site, unless additional information is obtained by ATSDR which, in the Agency's opinion, indicates a need to revise or append the conclusions previously issued.

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Prepared by:

**Exposure Investigation and Consultation Branch
Division of Health Assessment and Consultation
Agency for Toxic Substances and Disease Registry**

Background

The Herculaneum lead smelter is an active facility that has been operating since 1892. The Doe Run Company currently owns and operates the smelter. The facility is located at 881 Main Street in Herculaneum, Missouri. A lead ore concentrate, consisting of 80% lead sulfide, is processed at the smelter. The ore is obtained from lead mines operated by the company in the Viburnum Trend [1].

The company disposes waste slag from the smelting operations in a waste management area (WMA) south of the smelter. The WMA currently occupies about 24 acres of plant property. A large portion of the slag storage area is designated as a wetland and is subject to flooding from Joachim Creek and the Mississippi River.

Eight air monitoring stations in the surrounding area monitor for lead. Recent air monitoring data from all but one of these stations has been in compliance with the National Ambient Air Quality Standard (NAAQS) lead standard. The Broad Street station, which is located across the street from the smelter, has consistently documented lead concentrations in air that exceed the NAAQS for lead. Potential sources of lead emissions from the smelter facility include air emissions from the smelter stack, as well as fugitive dusts from the buildings and waste slag piles.

The Missouri Department of Health (MDOH) offers blood lead testing to residents of the area. Between June 1992 and May 1999, 52 children who live in the vicinity of the smelter were tested. Of these 52 children, 15 had blood lead concentrations above 10 $\mu\text{g/dL}$, and 3 had blood lead concentrations greater than 20 $\mu\text{g/dL}$ [2].

Staff from the MDOH have met with parents of children with elevated blood lead concentrations and counseled them on how to reduce exposure to environmental sources of lead. However, in spite of home inspections and personal counseling on how to reduce exposures to lead, some of the children continue to have elevated blood lead concentrations.

Therefore, the Agency for Toxic Substances and Disease Registry (ATSDR) worked with the MDOH to conduct this Exposure Investigation (EI). Staff from the MDOH identified homes occupied by children who had persistently elevated blood lead concentrations. Environmental and biological samples were collected from these homes and analyzed for total lead content, as well as for stable lead isotopes (lead 204, 206, 207, 208). These data, in conjunction with information from interviews with family members, were used to identify possible sources of lead exposure in the children with elevated blood lead concentrations.

This investigation was conducted as a public health service to the participants to assist them in better understanding their potential exposure to environmental lead contamination. The results obtained are applicable only to the participants of this investigation and may not be generalizable to other individuals or populations.

Target Population

ATSDR consulted with the MDOH to identify appropriate houses for testing. This Exposure Investigation (EI) was limited to two houses in Herculanum, which met the following criteria:

- (1) The house is located within a 1/2-mile radius of the smelter.
- (2) One or more children aged 6 years old or less has lived at the house for his/her entire life.
- (3) Recent testing has identified at least one child in the house who has a blood lead concentration of 15 µg/dL or higher.

In addition to testing the index child/children, other family members were also included in the investigation.

Test Procedures

Prior to testing, each participant, and a parent or legal guardian of each minor participant, was required to sign an informed consent/assent form. Written, informed consent was also obtained for environmental testing.

A registered nurse from the Jefferson County Health Department collected a blood sample from each family member using two 5-ml heparinized Vacutainers®. Each participant also provided a first morning void urine sample. The biological samples were stored on ice packs until they were shipped by overnight express delivery to the laboratory for analysis.

A representative of ATSDR collected the following environmental samples at each house:

- (1) A house dust sample was collected using a Nilfisk® HEPA vacuum cleaner from two rooms in the house that are frequented by the child.
- (2) A composite surface soil sample (0-1 inch) was collected along the drip line of the house on the side facing the smelter. A second composite surface soil sample was collected in a play area of the yard.
- (3) Paint samples were collected from surfaces identified by previous XRF surveys as having been painted with lead-containing paint.
- (4) A sample of first morning draw tap water was collected from the kitchen tap.
- (5) Two ambient air particulate samples were collected at each house using a 37 mm (0.8 µ pore size) mixed cellulose ester filter and an air pump that operated at a flow rate of 3 to 5 liters per minute. Air samples were collected 4-5 feet from the ground in an open area. ATSDR attempted to collect two 24-hour samples at each house, but the second sampling period was cut short because of threatening rain. Blank air filters were also analyzed for lead.

- (6) Hand wipes from both hands were collected from each child using pre-wetted baby wipes. After collection, the wipes were placed in pre-cleaned glass jars. Blank wipes were also analyzed for lead.

In addition, the Missouri Department of Natural Resources (MDNR) provided ATSDR with two air particulate filters from an air monitoring unit located at Dunklin High School. The filters were from 24-hour sampling events conducted on February 24, 2001 and March 14, 2001.

Analytical Procedures

The environmental and biological samples were shipped by overnight express delivery to the University of Texas at Dallas, Department of Geology, for analysis.

The samples were digested in concentrated nitric acid in a microwave oven. Aliquots of the digests were tested for total lead content using atomic absorption. Isotopic lead analysis for lead 204, 206, 207, and 208 were conducted using thermal ionization mass spectrometry [3]. Field blank samples of hand wipes, air filters, and vacutainers were also tested for lead. Isotopic lead standards (NIST SRM 981) were simultaneously run to confirm the accuracy and precision of the isotopic analyses.

Results

The lead concentrations and lead isotopic ratios for the biological and environmental samples collected from the two families that participated in this investigation are presented in Tables 1 and 2. The analytical results for two air samples collected by the MDNR at Dunklin High School are presented in Table 3.

To help discern the differences in the isotopic ratios, the lead ratios are presented graphically in Figures 1, 2, and 3. In these figures, only the 206 to 207 and the 206 to 204 ratios are presented. These ratios were selected because they have the widest range of variability [4]. It would be redundant to analyze the other ratios, since all the isotopic ratios strongly co-vary.

As indicated, the blood lead concentrations in the younger children (2 to 4 years old) are higher than those in their teenage siblings or parents. In the younger children, the blood lead concentrations range from 14.4 - 26.1 $\mu\text{g/dL}$, whereas the blood lead concentrations in teenagers and adults range from 3.2 - 10.9 $\mu\text{g/dL}$.

The data in Tables 1 and 2 show that the lead isotopic ratios are very similar in the blood and urine samples from an individual. This is to be expected, since lead that is removed from the blood by the kidneys is excreted into the urine. Other researches have noted that the lead isotopic ratio in blood and urine are very similar, whereas the lead mass concentrations are only weakly related [5].

The ambient air monitoring data indicate that elevated concentrations of lead were detected in air particulate samples collected in the EI participants' yards. On the first day of sampling, the air samples were collected over a time period of about 24 hours. On the second day of sampling, the air samples were collected over a time period of about 12 hours; the sampling period on the second day was cut short to protect the air sampling pumps from threatening rain. The lead concentrations in the residential air samples ranged from 0.73 to 6.3 $\mu\text{g}/\text{m}^3$. By comparison, the National Ambient Air Quality Standard for lead is 1.5 $\mu\text{g}/\text{m}^3$, as averaged over a three-month period.

The MDNR provided ATSDR with two air filters collected from a high volume air sampler located at Dunklin High School. The samples were selected to represent a maximum air lead concentration and a typical air lead concentration detected during a 24-hour sampling period. MDNR reported the lead concentrations in these two air samples to be 41.7 $\mu\text{g}/\text{m}^3$ and 4.14 $\mu\text{g}/\text{m}^3$, respectively.

At each house, a wipe sample was collected from an outdoor window sill. As indicated, the window sill wipe samples contained a high surface loading concentration of lead, and the lead isotopic ratios were similar to those in the air samples. This is to be expected, since a likely source of window sill dust is from the fall-out of air particulates.

Soil samples from House 2 contained high concentrations of lead (1,630 ppm and 2,900 ppm). In 1999, the yard soil from House 1 was removed and replaced with clean soil. A yard soil sample from House 1 contained 342 ppm lead, and a soil sample collected along the drip line of the house contained 999 ppm lead. An XRF survey did not detect leaded paint on the exterior of the house. Therefore, a possible source of the lead contamination at the drip line was air particulates that had deposited on the roof, which were then washed by rain onto the ground.

Discussion

The Centers for Disease Control determined that adverse health effects may occur in children (≤ 6 years old) at blood lead levels in excess of 10 $\mu\text{g}/\text{dL}$. As indicated by the data in Tables 1 and 2, the blood lead concentrations in the three young children in this EI exceeded this level of health concern.

In order to identify possible sources of lead exposure in these children, ATSDR conducted isotopic lead analyses. Lead from different geological sources has different isotopic ratios. By comparing the lead isotopic ratios in the blood samples with the isotopic ratios in potential environmental sources, it is sometimes possible to identify the source of a child's exposure.

As discussed previously, the blood lead concentrations in the children were higher than those in the adults. Furthermore, the data in Tables 1 and 2 show that the lead isotopic ratios in blood from the young children were different than the isotopic ratios in their teenage siblings or parents. This indicates that the lead exposures in children are quantitatively and qualitatively different than those in adults.

The concentrations of lead in the first-draw tap water samples were low. Furthermore, the lead isotopic ratios in water were far below those of any of the children's blood samples. Therefore, lead from tap water is not likely to be a significant source of exposure.

In House 1, the doorframe of one child's bedroom had been painted with lead-containing paint. However, this paint was in good condition, and it had been sealed with a new coat of paint. Furthermore, the lead isotopic ratio of this paint was considerably lower than the children's blood lead isotopic ratios, so this paint is not a likely source of exposure.

In House 2, lead-containing paint was identified on the walls of a storage area off of the garage. This area was inaccessible to the child. Furthermore, the lead isotopic ratio in this paint sample was considerably higher than the child's blood sample. Therefore, it is not likely that this paint is a source of the child's lead exposure.

Other potential sources of lead exposure in the environment of the children in this EI are the high levels of lead detected in air, dust, and soil. The isotopic lead ratios in these environmental media cluster near, but do not precisely match, the blood isotopic ratios. Therefore, rather than being exposed to a single environmental source, lead from varying combinations of these media may be contributing to the children's lead exposure.

Ambient air monitoring data have documented that air in the vicinity of the smelter has consistently exceeded the NAAQS for lead. Lead emissions from smelters is of particular health concern because of the high bioavailability of lead in smelter emissions [6]. The small particle size of the air particulates facilitate lead absorption if the particles are inhaled or ingested. In addition, children absorb a higher percent of lead from their gastrointestinal tracts than do adults [7].

Soil lead contamination within 1/4 -mile of the smelter averages about 3,000 ppm, whereas background soil lead concentrations in the area are 25 to 40 ppm [2]. Therefore, it is likely that the fall-out of lead-containing air particulates from the smelter is contributing to lead contamination in soil and window sill dust. These media could, in turn, be contributing to lead contamination in house dust.

It has been well-documented that lead contamination in surface soils and house dust is a significant source of lead exposure in young children [8]. Infants commonly exhibit pica activity (the ingestion of soil and other non-food items). In addition, young children often put their fingers and other soiled objects in their mouths, and this could result in the ingestion of contaminated soil and dust.

As shown in Figures 1, 2, and 3, the lead isotopic ratios in the children's hand wipe and house dust samples plotted near the children's blood isotopic ratios. Therefore, it is reasonable to speculate that the children could pick up contaminated house dust on their hands, which they then transfer to their mouths.

It is likely that the children in this EI were also exposed to small amounts of lead through their diet. Foods were not analyzed in this EI, since dietary intake of lead could vary widely from day to day. However, it is not likely that lead in food is a major source of lead exposure, since the dietary intake of lead by a 2-year old child is estimated to be only 4.3 $\mu\text{g/day}$ - a relatively small dose [7]. Furthermore, blood lead levels in other family members, who presumably eat many of the same foods as the children, were not elevated.

Since the young children in this EI have lived their entire lives at their current residences, they have not had the opportunity for significant exposures to environmental lead in other geographical areas. Furthermore, none of the children had been breast fed, so they were not exposed to lead in their mother's breast milk.

It is possible that another unidentified source of lead is contributing to the elevated blood lead levels in these children. The lead isotope ratios in many of the air, soil, and house dust samples slightly exceeded the lead isotope ratios in the blood samples. Therefore, it is possible that another unidentified lead source, particularly one with a lead isotope ratio lower than those in the children's bloods, is contributing to their lead exposure. However, the comprehensive environmental screening conducted during this EI and detailed interviews with the families failed to identify other potential sources of lead exposure.

Conclusions

- (1) Blood lead concentrations were elevated above a level of health concern in young children (2 to 4 years old).
- (2) Lead contamination was detected in ambient air particulates, dust on window sills, surface soil, and house dust. Lead from these sources is likely contributing to the elevated blood lead concentrations detected in the young children.
- (3) Lead in paint and water do not appear to be a significant source of lead exposure in the children with elevated blood lead concentrations.

Recommendations

- (1) Implement actions to minimize children's exposures to lead-contaminated soil and house dust.
- (2) Implement actions to reduce lead concentrations in ambient air.
- (3) Encourage regular blood lead testing in all young children (≤ 6 years old) who live in the vicinity of the smelter.

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- (2) U. S. Environmental Protection Agency; Administrative Order of Consent; Docket No. RCRA-7-2000-0018; September 29, 2000.
- (3) W. I. Manton et al.; Acquisition and retention of lead by young children; Environmental Research, Section A; 82 60-80 (2000).
- (4) M. B. Rabinowitz; Stable isotopes of lead for source identification; Clinical Toxicology 33(6) 649-655 (1995).
- (5) B. L. Gulson et al.; Blood lead-urine lead relationships in adults and children, Environmental Research, Section A 78 152-160 (1998).
- (6) M. J. Steele et al.; Assessing the contribution from lead in mining wastes to blood lead; Regulatory Toxicol Pharmacol 11:158-190 (1990).
- (7) Agency for Toxic Substances and Disease Registry; Toxicological Profile for Lead (Update); July 1999.
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Table 1

House 1

Biological Samples

Age	Type	Lead	206/204	207/204	208/204	206/207
(mother)	Blood	4.2 µg/dL	19.69	15.64	38.77	1.259
	Urine	2.8 µg/L	19.81	15.73	39.02	1.260
(father)	Blood	10.9 µg/dL	19.60	15.77	39.01	1.242
	Urine	3.7 µg/L	19.52	15.72	38.89	1.243
16yr	Blood	7.2 µg/dL	19.70	15.65	38.83	1.259
	Urine	9.8 µg/L	19.87	15.78	39.16	1.259
4yr	Blood	15.9 µg/dL	20.14	15.77	39.26	1.277
	Urine	13.6 µg/L	20.20	15.80	39.33	1.278
	Handwipe 1	41 µg	20.29	15.83	39.43	1.282
	Handwipe 2	75 µg	20.40	15.85	39.56	1.287
2yr	Blood	26.1 µg/dL	20.17	15.80	39.32	1.276
	Urine	24.7 µg/L	20.16	15.78	39.28	1.278
	Handwipe 1	34 µg	19.63	15.76	38.94	1.246
	Handwipe 2	66 µg	20.35	15.81	39.46	1.287

Environmental Samples

Water	3.2 µg/L	18.75	15.66	38.45	1.197
Air 1	2.29 µg/m3	20.62	15.85	39.64	1.301
Air 2	0.733 µg/m3	20.67	15.88	39.72	1.302
House Dust 1	821 µg/m2	20.33	15.85	39.54	1.283
House Dust 2	1,796 µg/m2	20.44	15.82	39.55	1.291
Window sill	12,775 µg/m2	20.52	15.83	39.55	1.296
		20.55	15.86	39.65	1.297
Soil 1	342 µg/g	20.34	15.83	39.50	1.285
		20.34	15.82	39.48	1.285
Soil 2	999 µg/g	20.39	15.83	39.52	1.288
		20.38	15.82	39.48	1.288
Paint	17,300 µg/g	19.35	15.69	38.84	1.233
		19.33	15.68	38.59	1.233

Table 2

House 2

Age	Type	<i>Biological Samples</i>				
		Lead	206/204	207/204	208/204	208/207
(mother)	Blood	3.0 µg/dL	19.69	15.64	38.79	1.259
	Urine	6.1 µg/L	20.07	15.81	39.32	1.270
(father)	Blood	6.8 µg/dL	19.78	15.83	39.22	1.249
	Urine	4.35 µg/L	19.68	15.73	39.00	1.251
13yr	Blood	7.8 µg/dL	20.13	15.79	39.28	1.275
	Urine	7.8 µg/L	20.14	15.78	39.27	1.276
	Handwipe	348 µg	20.51	15.89	39.71	1.291
4yr	Blood	14.4 µg/dL	20.32	15.81	39.42	1.285
	Urine	20.1 µg/L	20.31	15.81	39.43	1.285
	Handwipe 1	239 µg	20.53	15.85	39.60	1.295
	Handwipe 2	96 µg	20.39	15.84	39.58	1.287

Environmental Samples

Water	0.18 µg/L	19.62	15.68	38.90	1.252
Air 1	3.28 µ/m3	19.83	15.76	39.50	1.258
Air 2	6.30 µ/m3	20.46	15.84	39.51	1.291
House Dust 1	723 µg/m2	20.31	15.84	39.48	1.283
House Dust 2	505 µg/m2	20.19	15.82	39.38	1.276
Window sill	48,100 µg	20.47	15.87	39.65	1.290
		20.42	15.82	39.47	1.291
Soil 1	1,630 µg/g	20.74	15.87	39.76	1.307
		20.74	15.85	39.71	1.308
Soil 2	2,900 µg/g	20.78	15.87	39.73	1.309
		20.77	15.86	39.69	1.310
Paint	6,490 µg/g	20.90	15.84	40.20	1.318
		20.89	15.83	40.12	1.319

Table 3

Ambient Air Samples at Dunklin High School (MDNR)

Date	Lead	206/204	207/204	208/204	208/207
2/24/01	41.7 $\mu\text{g}/\text{m}^3$	19.99	15.79	39.18	1.266
3/14/01	4.14 $\mu\text{g}/\text{m}^3$	20.21	15.82	39.36	1.278

Legend for Figures 1 - 3

Air HS - Air sample collected at Dunklin High School

HW - Hand Wipe from child

HD - House dust sample

WSD - Dust sample from outdoor window sill

Figure 1: Lead isotopic ratios in 4-year old boy

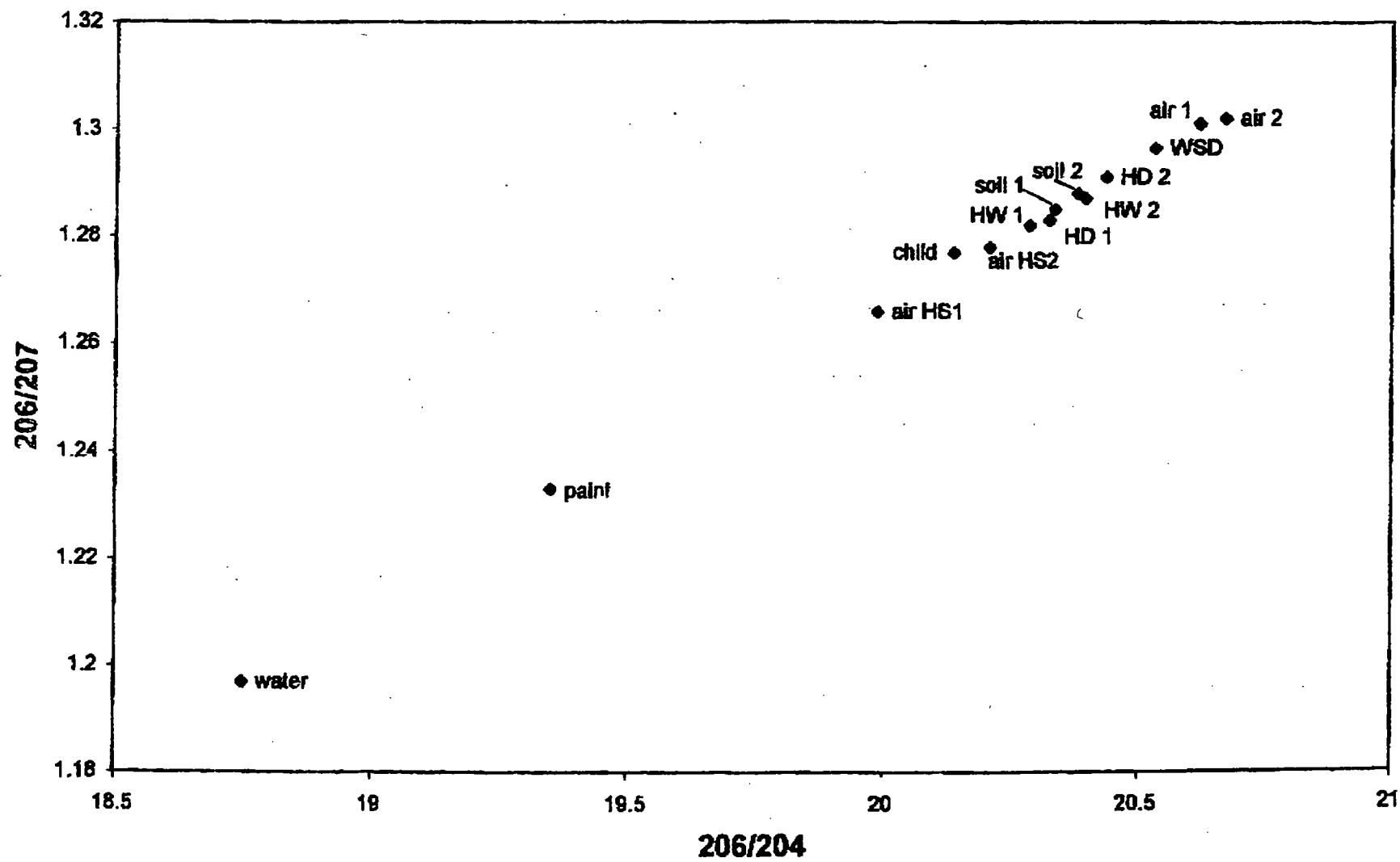


Figure 2: Lead isotopic ratios in 2-year old girl

